

(51) International Patent Classification <sup>6</sup> : B01J 2/00		A1	(11) International Publication Number: WO 97/07879
			(43) International Publication Date: 6 March 1997 (06.03.97)
(21) International Application Number: PCT/US96/13582		(81) Designated States: CA, JP, US, European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).	
(22) International Filing Date: 21 August 1996 (21.08.96)			
(30) Priority Data: 60/002,896 29 August 1995 (29.08.95) US		Published With international search report.	
(60) Parent Application or Grant (63) Related by Continuation US 60/002,896 (CON) Filed on 29 August 1995 (29.08.95)			
(71) Applicant (for all designated States except US): E.I. DU PONT DE NEMOURS AND COMPANY [US/US]; 1007 Market Street, Wilmington, DE 19898 (US).			
(72) Inventor; and (75) Inventor/Applicant (for US only): SCHURR, George, Alan [US/US]; 12 The Horseshoe, Newark, DE 19711-2069 (US).			
(74) Agent: HAMBY, Jane, O.; E.I. du Pont de Nemours and Company, Legal Patent Records Center, 1007 Market Street, Wilmington, DE 19898 (US).			

The diagram shows a fluid control system 10. A reservoir 20 is connected to a pump 18, which feeds into a valve assembly 12. The valve assembly has a main line 14 with a check valve 31 and a side branch 22 containing a solenoid 24. A second pump 26 also feeds into the side branch 22. A hopper 30 feeds material into a funnel 28, which discharges into the main line 14. The main line 14 leads to a container 32.

The process and apparatus of the present invention coat a solid particle by metering a liquid composition comprising a coating material, where the liquid composition is either a solution, a slurry or a melt, into a flow restrictor and injecting a gas stream through the flow restrictor concurrently with the metering of the liquid composition to create a zone of turbulence at the outlet of the flow restrictor, thereby atomizing the liquid composition. The gas stream is heated prior to injecting it through the flow restrictor. A solid particle is added to the zone of turbulence concurrently with the metering of the liquid composition and the injection of the heated gas to mix the solid particle with the atomized liquid composition. The mixing at the zone of turbulence coats the solid particle with the coating material. If the liquid composition is a melt, then the solid particle is coated with the melt. If the liquid composition is either a solution or a slurry, the solid particle is coated with either the dissolved or the undissolved solid of the liquid composition. This apparatus and process provide a short residence time, i.e., less than 250 milli-seconds, in the zone of turbulence, which reduces the time and thus the cost of coating particles. In addition, this process and apparatus provide a mechanism for coating very small or powdery or granular particles which results in a high yield of entirely coated, non-agglomerated particles.

**FOR THE PURPOSES OF INFORMATION ONLY**

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AM	Armenia	GB	United Kingdom	MW	Malawi
AT	Austria	GE	Georgia	MX	Mexico
AU	Australia	GN	Guinea	NE	Niger
BB	Barbados	GR	Greece	NL	Netherlands
BE	Belgium	HU	Hungary	NO	Norway
BF	Burkina Faso	IE	Ireland	NZ	New Zealand
BG	Bulgaria	IT	Italy	PL	Poland
BJ	Benin	JP	Japan	PT	Portugal
BR	Brazil	KE	Kenya	RO	Romania
BY	Belarus	KG	Kyrgyzstan	RU	Russian Federation
CA	Canada	KP	Democratic People's Republic of Korea	SD	Sudan
CF	Central African Republic	KR	Republic of Korea	SE	Sweden
CG	Congo	KZ	Kazakhstan	SG	Singapore
CH	Switzerland	LI	Liechtenstein	SI	Slovenia
CI	Côte d'Ivoire	LK	Sri Lanka	SK	Slovakia
CM	Cameroon	LR	Liberia	SN	Senegal
CN	China	LT	Lithuania	SZ	Swaziland
CS	Czechoslovakia	LU	Luxembourg	TD	Chad
CZ	Czech Republic	LV	Latvia	TG	Togo
DE	Germany	MC	Monaco	TJ	Tajikistan
DK	Denmark	MD	Republic of Moldova	TT	Trinidad and Tobago
EE	Estonia	MG	Madagascar	UA	Ukraine
ES	Spain	ML	Mali	UG	Uganda
FI	Finland	MN	Mongolia	US	United States of America
FR	France	MR	Mauritania	UZ	Uzbekistan
GA	Gabon			VN	Viet Nam

**TITLE****APPARATUS AND PROCESS FOR COATING A SOLID PARTICLE****BACKGROUND OF THE INVENTION****1. Field of the Invention**

5           This invention relates to an apparatus and a process for uniformly coating, or encapsulating, a solid particle. More specifically, this invention relates to an apparatus and a process for coating a solid particle with either a melt or a solid, by  
10       respectively metering a melt, a solution in which a solid is dissolved or a slurry in which a solid is undissolved.

**2. Description of the Related Art**

15           Methods and apparatus for coating particles which use heated gas are known in the art. For example, U.S. Patent No. 4,784,878 to Haak discloses a method of applying a coating of a non-volatile substance, such as lecithin, to a solid particle. In Haak, there is a concurrent flow of the atomized liquid/solid mixture,  
20       hot gas and an atomized liquid containing lecithin. The atomized liquid containing liquid ultimately comes into contact with the solid particle of the liquid/solid mixture, and exterior coating of the solid particle with lecithin occurs. Thus, the solid which  
25       was part of the mixture is coated. This method is inefficient in that it requires the particles which are coated to be mixed with a liquid and the liquid to be subsequently evaporated in order to recover the particles.

30           It is especially difficult to coat very small particles, such as a powdery or granular material. A method using heated gas for coating such particles is described in U.S. Patent No. 5,075,138 to Tanaka et al. In this patent, hot air is supplied to a region spaced  
35       in the downstream direction from a region where the powdery or granular material and a coating liquid collide with each other. With this method, the mechanism for transforming the coating liquid to a solid coating is not particularly efficient, since most

of the coating liquid ends up in the liquid phase, and not in the solid phase as the coating on the powdery or granular material. Consequently, the resultant yield of coated powdery or granular material is not high, and both coating liquid and the powdery or granular material are wasted.

Mixing one material with a particulate material in a zone of turbulence is disclosed in U.S. Patent No. 4,430,001 to Schurr. However, the one material is 0.05%-6% by weight based on the mixture, and thus does not encapsulate the particulate material.

Thus, there exists a need for developing an apparatus and a process for coating solid particles, and in particular, small particles, such as powdery or granular materials, which apparatus and process are efficient and economical.

#### SUMMARY OF THE INVENTION

The present invention solves the problems of the prior art associated with uniformly and completely coating, or encapsulating, a solid particle. In particular, the apparatus and the process of the present invention provide a mechanism for coating particles which requires only a short residence time in a mixing area, or zone of turbulence. This reduces the time and hence, the cost, of coating particles.

In addition, the present invention provides a mechanism for coating small particles, such as powdery or granular materials, where the yield of uniformly and completely coated particles is high. Thus, little of the coating material and the solid particles is wasted, which reduces the cost of coating particles.

Moreover, the present invention provides methods for coating a solid particle with either a melt or a solid, by respectively metering a melt, a solution in which a solid is dissolved, or a slurry, in which a solid is undissolved. This allows a variety of solid particles to be coated with a variety of materials, thereby exceeding the versatility of known coating methods of the prior art.

To achieve the objects and in accordance with purposes of the invention, as embodied and broadly described herein, there is provided an apparatus for coating a solid particle with a coating material, the apparatus comprising a first chamber; a flow restrictor disposed at one end of the chamber; a liquid inlet line disposed in fluid communication with the chamber for metering a liquid composition into the chamber and through the flow restrictor; a gas inlet line disposed in fluid communication with the first chamber for injecting a first gas stream through the flow restrictor to create a zone of turbulence; and means disposed in the gas inlet line and upstream of the flow restrictor for heating the first gas stream prior to injection through the flow restrictor.

Further in accordance with the present invention, there is provided a process for coating a solid particle with a coating material. The process comprises the steps of metering a liquid composition comprising the coating material into a flow restrictor having an outlet end; injecting a gas stream through the flow restrictor concurrently with the metering step to create a zone of turbulence at the outlet end of the flow restrictor, thereby atomizing the liquid composition; heating the gas stream prior to injecting the gas stream through the flow restrictor; and adding a solid particle to the zone of turbulence concurrently with the metering and injecting steps to mix the solid particle with the atomized liquid composition, wherein the mixing at the zone of turbulence coats the solid particle with the coating material.

The liquid composition may be a solution, in which the coating material is dissolved in a liquid or a slurry, in which the coating material is not dissolved in a liquid. Alternatively, the liquid composition may comprise a melt.

#### **BRIEF DESCRIPTION OF THE DRAWINGS**

Fig. 1 is a schematic diagram of a portion of the apparatus in accordance with the present invention.

Fig. 2 is a cut-away, expanded, cross-sectional view of a portion of the apparatus shown in Fig. 1.

**DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS**

Reference will not be made in detail to the present preferred embodiments of the invention, an example of which is illustrated in the accompanying drawings. Wherever possible, the same reference numbers will be used throughout the drawings to refer to the same or like parts.

The present invention provides for an apparatus for coating a solid particle with a coating material. By coating is meant adhering a layer of one substance onto the surface of a solid particle, and includes encapsulation of substantially all, or all, the surface of the solid particle.

An apparatus according to the present invention is shown generally at 10 in Fig. 1. The apparatus of the present invention comprises a first chamber, shown at 12 in Figs. 1 and 2. A flow restrictor 14 is disposed at one end of the first chamber. The flow restrictor is typically disposed at the downstream end of the first chamber, as shown in Figs. 1 and 2. Flow restrictor 14 has an outlet end 14a, as shown in the detailed view of Fig. 2. Although the flow restrictor is shown as a different element from the first chamber, it may be formed integrally therewith, if desired. The flow restrictor of the present invention may have various configurations, as long as it serves to restrict flow and thereby increase the pressure of the fluid passing through it. Typically, the flow restrictor of the present invention is a nozzle.

A first, or liquid, inlet line 16 as shown in Figs. 1 and 2 is disposed in fluid communication with the first chamber for metering a liquid composition into the chamber. Liquid inlet line 16 meters the liquid composition into first chamber 12 in the outlet of flow restrictor 14, and preferably in the center of the flow restrictor when viewed along the axial length thereof. The liquid composition is metered through

liquid inlet line 16 by a metering pump 18 from a storage container 20 containing the liquid composition as shown in Fig. 1. The liquid composition may be a solution, where a solid which is used as the coating material is dissolved in a liquid, or a slurry, where a solid which is used as the coating material is undissolved in a liquid. Alternatively, the liquid composition may be a melt, which is used as the coating material. By melt is meant any substance at a temperature at or above its melting point, but below its boiling point. In any of these cases, the liquid composition may include components other than the coating material. It should be noted that when the liquid composition is a melt, storage container 20 must be heated to a temperature above the melt temperature of the liquid composition in order to maintain the liquid composition in melt form.

The apparatus for coating a solid particle further includes a second, or gas, inlet line 22 disposed in fluid communication with the first chamber as shown in Figs. 1 and 2. Generally, the gas inlet line should be disposed in fluid communication with the first chamber upstream of the flow restrictor. Gas inlet line 22 injects a first gas stream through the flow restrictor to create a zone of turbulence at the outlet of the flow restrictor. The turbulence subjects the liquid composition to shear forces that atomize the liquid composition.

The first gas stream should have a stagnation pressure sufficient to accelerate the gas to at least one-half the velocity of sound, or greater, prior to entering the flow restrictor to ensure that a zone of turbulence of sufficient intensity will be formed at the outlet of the flow restrictor. The velocity of sound for a particular gas stream, e.g., air or nitrogen, will be dependent on the temperature of the gas stream. This is expressed by the equation for the speed of sound,  $c$ :

$$c = \sqrt{kgRT} \quad (1)$$

where:

k = ratio of specific heats for the gas

5 g = acceleration of gravity

R = universal gas constant

T = absolute temperature of the gas

Thus, the acceleration of the first gas stream is dependent on the temperature of the gas stream.

10 As noted above, it is the pressurized gas that causes the atomization of the liquid composition. The pressure of the liquid composition in the liquid inlet line just needs to be enough to overcome the system pressure of the gas stream. It is preferable that the  
15 liquid inlet line has an extended axial length before the zone of turbulence. If the liquid inlet line is too short, the flow restrictor becomes plugged.

The apparatus of the present invention also comprises means disposed in the second inlet line and  
20 upstream of the flow restrictor for heating the first gas stream prior to injection through the flow restrictor. Preferably, the heating means comprises a heater 24 as shown in Fig. 1. Alternatively, the heating means may comprise a heat exchanger, a  
25 resistance heater, an electric heater, or any type of heating device. Heater 24 is disposed in second inlet line 22. A pump 26 as shown in Fig. 1 conveys the first gas stream through heater 24 and into first chamber 12. When a melt is used as the coating  
30 material, the gas stream should be heated to a temperature at or above the melt temperature of the melt, to keep the melt in liquid (i.e., melt) form. When using a melt, it is also helpful if auxiliary heat is provided to the first inlet line which supplies the  
35 melt prior to injection, to prevent pluggage of the line.

The apparatus of the present invention further includes a hopper 28 as shown in Figs. 1 and 2. Hopper 28 introduces a solid particle to the zone of



turbulence. It is preferable that the outlet end of the flow restrictor is positioned in the first chamber beneath the hopper at the center line of the hopper. This serves to ensure that the solid particles are

5 introduced directly into the zone of turbulence. This is important because, as noted above, the turbulence subjects the liquid composition to shear forces that atomize the liquid composition. It also increases operability by providing a configuration for feeding

10 the solid particles most easily. In addition, the shear forces disperse and mix the atomized liquid composition with the solid particles, which allows the particles to be coated. Hopper 28 may be fed directly from a storage container 30 as shown by arrow 29 in

15 Fig. 1. The hopper of the present invention may include a metering device for accurately metering the particles at a particular ratio to the liquid feed from liquid inlet line 16 into the zone of turbulence. This metering establishes the level of coating on the solid

20 particle. Typically, the hopper of the present invention is open to the atmosphere. When a melt is used, it is preferred that the solid particles are at ambient temperature because this facilitates solidification of the melt after the melt, which is

25 initially at a higher temperature, coats the solid particle in the zone of turbulence.

The apparatus of the present invention may further include a second chamber 32 surrounding the first chamber as shown in Figs. 1 and 2. In addition,

30 the second chamber encloses the zone of turbulence. Second chamber 32 has an inlet 34 for introducing a second gas stream into the second chamber. The inlet of the second chamber is preferably positioned at or near the upstream end of second chamber 32. The outlet

35 of second chamber 32 is connected to a collection container, such as that shown at 36 in Fig. 1. The second gas stream cools and conveys the coated solid particles toward the collection container as illustrated by arrow 31 in Fig. 2. In particular, when

a solution or slurry is used, the solid of the solution or slurry cools between the zone of turbulence and container so that by the time the particle reaches the container, a solid coating comprising the solid of the solution or slurry is formed on the particle. When a melt is used, the liquid composition cools between the zone of turbulence so that by the time the particle reaches the container, a solid coating comprising the melt is formed on the particle. The first gas stream, as well as the second gas stream, are vented through the top of collection container 36.

For the configuration as shown in Figs. 1 and 2, inlet 34 may be connected to a blower, not shown, which supplies the second gas stream to the second chamber. However, the blower and second chamber 32 may be eliminated, and the first gas stream may be used to cool the particles and to convey them to container 36. In this case, the solid from the solution or slurry or the melt cools and solidifies on the particle in the atmosphere between the zone of turbulence and the collection container, and the coated particles fall into collection container 36.

It is preferable that the axial length of the zone of turbulence is about ten times the diameter of the second chamber. This allows the pressure at the outlet of the flow restrictor to be at a minimum. Solid particles are fed into second chamber 32 as shown in Figs. 1 and 2 near the outlet of the flow restrictor, which is preferably positioned at the center line of the hopper. If the pressure at the outlet is too great, the solid particles will back flow into the hopper.

The pressure of the second gas stream must be sufficient to assist in conveying the coated solid particles from the zone of turbulence to the collection zone, but should be at lower than the pressure of the first gas stream. This is because a high relative velocity difference between the first gas stream and

the second gas stream produces a sufficient degree of turbulence to coat the solid particles.

Further in accordance with the present invention, there is provided a process for coating a solid  
5 particle with a coating material. It should be noted that the process of the present invention may be practiced using the apparatus illustrated in Figs. 1 and 2, although it should be understood that the process of the present invention is not limited to the  
10 illustrated apparatus. Moreover, it should be noted that while one pass, or cycle, of the process of the present invention completely coats, or encapsulates, the solid particle, more than one pass may be used to adhere additional coating material to the solid  
15 particle, depending on the desired thickness of the coating.

The process comprises the steps of metering a liquid composition into a flow restrictor, such as flow restrictor 14 as shown in Figs. 1 and 2. As described  
20 above for the apparatus, the liquid composition may be a solution, slurry or melt.

The process of the present invention further comprises injecting a gas stream, for instance from a gas inlet line such as that shown at 22 in Figs. 1 and  
25 2, through the flow restrictor concurrently with metering the liquid composition into the flow restrictor, to create a zone of turbulence at the outlet of the flow restrictor. The shear in the zone of turbulence atomizes the liquid composition.

30 The gas stream is heated prior to injecting it through the flow restrictor. The gas stream may be heated by a heater, such as heater 24 as shown in Fig. 1. As noted above for the apparatus, when the liquid composition is a solution or a slurry, the gas  
35 stream is heated to a temperature sufficient to vaporize the liquid of the solution or slurry and to leave the solid of the solution or slurry remaining. When the liquid composition is a melt, the gas stream should be heated to a temperature at or above the melt

temperature of the liquid composition, to keep the liquid composition, and in particular, the melt, in liquid (i.e., melt) form. As also noted above for the apparatus, when using a melt, it is also helpful if  
5 auxiliary heat is provided to the first inlet line which supplies the melt prior to injection, to prevent pluggage of the line.

The process of the present invention also comprises the step of adding a solid particle to the  
10 zone of turbulence concurrently with the metering of the liquid composition and the injection of the gas stream. This mixes the solid particle with the atomized liquid composition at the zone of turbulence. This mixing at the zone of turbulence coats the solid  
15 particle with the coating material. The solid is preferably metered in order to control the ratio of the solid and the liquid added at the zone of turbulence. This establishes the level of coating on the solid particle. When a solution or slurry is used, the heat  
20 from the heated gas stream serves to evaporate the liquid of the solution or slurry, leaving the solid of the solution or slurry remaining to coat the particle. The mixing at the zone of turbulence then coats the solid particle with the remaining solid from the  
25 solution or slurry. When a melt is used, the mixing at the zone of turbulence coats the solid particle with the melt.

As noted above, the zone of turbulence is formed by the action of injecting the gas at high pressure  
30 through the flow restrictor. As discussed above with respect to the apparatus, it is preferable that the gas stream is accelerated to at least about one-half the velocity of sound prior to injection to ensure that a zone of turbulence of sufficient intensity will be  
35 formed at the outlet of the flow restrictor.

The residence time of the particles in the zone of turbulence is determined by the geometry of the first chamber and the amount of gas injected from the gas inlet line. The average residence time of the

solid particle within the zone of turbulence is preferably less than 250 milli-seconds. More preferably, the average residence time of the solid particle within the zone of turbulence is in the range of 25 to 250 milli-seconds. Short residence times can be achieved because of the action of the zone of turbulence. The short residence times make the process of the present invention advantageous compared to conventional coating processes because the time, and hence, the cost of coating particles, are reduced.

Typically, the solid particles are fed from a hopper, such as hopper 28 as shown in Figs. 1 and 2, which is open to the atmosphere. As noted above for the apparatus, when the liquid composition is a melt, it is preferred that the solid particles be at ambient temperature because this will facilitate solidification of the melt after the melt (which is initially at a higher temperature) coats the solid particle in the zone of turbulence.

The process of the present invention may further comprise the step of adding another gas stream upstream of the zone of turbulence for cooling and conveying the coated solid particle. This other gas stream is added through a chamber, such as second chamber 32 as shown in Figs. 1 and 2. As explained above for the apparatus, the pressure of the second gas stream must be sufficient to assist in conveying the coated solid particles from the zone of turbulence to the collection container, but should be at lower than the pressure of the first gas stream in order to achieve coating. When a solution or slurry is used, the solid of the solution or slurry cools and solidifies on the particle in the second chamber between the zone of turbulence and a collection container, such as collection zone 36 as described above. When a melt is used, the melt cools and solidifies on the particle in the second chamber between the zone of turbulence and the collection container. When a second chamber is not included, the solid or the melt cools and solidifies on the particle

in the atmosphere between the zone of turbulence and the collection container, and the coated particles fall into the container.

The present invention will be clarified by the following Examples, which are intended to be purely exemplary of the invention.

#### EXAMPLE 1

Dodecanedioic acid is a chemical intermediate which has a tendency to sublime at elevated temperatures. In order to inhibit this sublimation, DDDA was encapsulated with a glycidyl methacrylate, or GMA, co-polymer. Specifically, this Example is directed to the encapsulation of solid particle of dodecanedioic acid (DDDA) with a GMA co-polymer of glycidyl methacrylate, butyl methacrylate, methyl methacrylate and styrene, which co-polymer is available from Anderson Development Company of Adrian, Michigan. The uncoated dodecanedioic acid solid particle had a mean particle size of 23.5 micrometers ( $\mu$ ), and the coated dodecanedioic acid had a mean particle size of 26.2 micrometers ( $\mu$ ).

Injector technology apparatus, such as that shown in Figs. 1 and 2, was used in order to encapsulate the DDDA with GMA co-polymer. The apparatus had a shear zone of one inch (2.54 cm.) in diameter. DDDA was dispersed using an Alpine 160z pin mill operated at 10,400 rpm to produce particles having a mean particle size of 23.5 micrometers ( $\mu$ ). DDDA powder was metered using a screw feeder at a rate of 1300 g./min. to the hopper as shown in Figs. 1 and 2. A solution comprising 43% by weight of the GMA co-polymer as described above, dissolved in toluene, was metered with a peristaltic pump at a rate of 242 g./min. A first gas stream of nitrogen at a pressure of 100 psig and at a temperature of 240° C was injected through the flow restrictor. The GMA co-polymer in toluene solution coated the DDDA particles in the zone of turbulence simultaneously with the flashing of the toluene solution to the gaseous phase as it was contacted by

the heated nitrogen gas stream. The mean residence time in the zone of turbulence was 150 milli-seconds.

The DDDA particles were collected and found to be substantially encapsulated by the GMA co-polymer. The particles had an average coating of 8% by weight of the GMA co-polymer, i.e., the coating was 8% of the total weight of the coated particle. Total feed of the coating material (i.e., 43% by weight of the GMA co-polymer described above, dissolved in toluene) was 18.6% based upon the weight of the DDDA.

The collected sample of coated DDDA particles was incorporated into a standard powder coating test formulation, coated onto a metal body panel, and then baked in a curing oven equipped with a collection device to measure volatiles by head space gas chromatography. The sample yielded reductions in DDDA volatiles of 80% by weight. The sample gave a cured coating with good gloss, no pitting, and good flow characteristics during curing in the bake oven.

Additional samples were prepared and tested in the manner described above. Particles encapsulated with an average coating of 11.1% by weight of the GMA co-polymer yielded a reduction in DDDA volatiles of 85%. Particles encapsulated with an average coating of 4.7% by weight of the GMA co-polymer yielded reduction in DDDA volatiles of 50%.

#### EXAMPLE 2

This Example is directed to encapsulating calcium carbonate with wood rosin. Calcium carbonate powder with a mean particle size of 10 micrometers ( $\mu$ ) was coated with wood rosin using the apparatus as shown in Figs. 1 and 2, and as described above.

Wood rosin, having a melting point of 120° C, was heated to 140° C and held at this temperature in an insulated bath. This wood rosin was metered with a peristaltic pump at a rate of 681 g/min. The atomizing air was heated to 140° C and used at 100 psig.

The calcium carbonate was metered in a screw feeder at a rate of 11,340 g/min to the hopper of the

apparatus as shown in Figs. 1 and 2. The wood rosin encapsulated the calcium carbonate particle at a coating level of 6%.

Uncoated calcium carbonate exposed to HCl will  
5 produce carbon dioxide gas due to chemical reaction. Encapsulation was proven by placing the encapsulated particles in a bath of hydrochloric acid (HCl). There was no immediate effervescence from the calcium carbonate indicating the complete surface protection  
10 provided by the wood rosin.

### EXAMPLE 3

This Example is directed to a sulfonylurea herbicide coated with either wax or rosin coatings, which were metered in the form of a melt. In this  
15 Example, Technical L5300 (tribenuron methyl) sulfonylurea herbicide, having a particle size ranging from about 2 to about 40 micrometers ( $\mu$ ), was metered in a screw feeder at a rate of 600 g./min.

Sulfonylurea herbicide was coated with rosin at  
20 190° C, metered with a gear pump at a rate of 75.6 g./min., corresponding to a coating level of 12.6% rosin, i.e., the weight of the rosin coating was 12.6% of the weight of the coated particle. A second batch of sulfonylurea herbicide was coated with wax at  
25 110° C, metered with a peristaltic pump at a rate of 56.4 g./min., corresponding to a coating level of 9.4% wax. The wax was a mixture of a partially oxidized, low molecular weight polyethylene (EPOLENE E10) and paraffin. A first gas stream comprising nitrogen was  
30 injected through a flow restrictor, where the gas stream was at a pressure of 38-40 psi just before the flow restrictor.

Sulfonylurea herbicides are normally degraded in the presence of hormonal/phenoxy herbicides, such as  
35 sodium 2,4-D and 2,4-D acid. Thus, the effectiveness of the coatings to inhibit this degradation was tested by measuring the degradation of sulfonylurea herbicide when mixed in 1/20 weight mixtures with sodium 2,4-D and 2,4-D acid and aged for three weeks at 45° C.



Uncoated sulfonylurea herbicide was tested in the same manner to provide a control. The results were as follows:

## PERCENTAGE RELATIVE DEGRADATION OF L5300

	<u>Uncoated</u>	12.6% Rosin <u>Coating</u>	9.4% Wax <u>Coating</u>
Sodium 2,4-D	38.4%	0.0%	0.0%
2,4-D Acid	21.2%	0.0%	1.7%

5

Thus, the results show coating at a level of 12.6% by weight essentially resulted in encapsulation of the particles. Generally, degradation of 5% or less is suitable for such formulations.

10 Additionally, the wax and rosin coated sulfonylurea was mixed with an ethoxylated silicone surfactant and water, and was then sprayed onto wild buckwheat, spring wheat and wild mustard. Both efficacy for weed injury and safety for the spring wheat was equivalent  
15 for the coated and uncoated sulfonylurea herbicides.

EXAMPLE 4

This Example is directed to a relatively water soluble herbicide, bromacil (5-bromo-3-sec-butyl-6-methyluracil) (IUPAC), having 815 ppm. water  
20 solubility, coated with stearic acid. In this Example, technical solid particles of bromacil, having a particle size range from less than one micrometer to 30 micrometers maximum diameter, was metered in a screw feeder at a rate of 1000 g./min.

25 The bromacil was coated with stearic acid in the form of a melt at a rate of 60 g./min. The stearic acid, which had a melting point of 70° C. was heated to 90° C and held at this temperature in a storage container, such as storage container 20 as described  
30 above. This first pass coated material was collected and coated a second time, corresponding to a coating level of 6% and 12% coating level on the first pass and second pass, respectively, although it should be noted that it is not necessary to coat the particles with

more than one pass in order to encapsulate the particles.

5 A gas stream comprising nitrogen was injected through a flow restrictor, such as that shown in Figs. 1 and 2. The gas stream was at a pressure of 70 psi just before the flow restrictor.

Particle size measurements were done using a Sympatec Helos. The Helos uses forward light scattering to measure particle size distribution. A well dispersed stream of dry particles is passed through a columnated laser beam. The resulting diffraction pattern is deconvoluted into a size distribution. Table 1 shows the particle size analysis of uncoated bromacil, first pass coated bromacil and second pass coated bromacil.

10

15

TABLE 1

Microns Diameter of Coated vs. Uncoated Bromacil

Particle Fraction	Uncoated Bromacil Zero Stearic Acid	First Pass 6% stearic acid	Second Pass 11.9% stearic acid
5% smaller than	0.7	0.8	0.9
10% smaller than	1.0	1.2	1.3
16% smaller than	1.3	1.8	1.9
50% smaller than	4.6	5.5	5.4
90% smaller than	12.0	20.0	19.3

Each of the size fractions of bromacil was coated, and the size analysis shows that the coated particles exist as individual discrete, non-agglomerated particles. The tiniest particles were still present, but with a slight increase in size as a result of being coated.

Additional advantages and modifications will readily occur to those skilled in the art. The invention, in its broader aspects, is therefore not limited to the specific details, representative apparatus and illustrative Examples shown and described. Accordingly, departures may be made from such details without departing from the spirit or scope of the general inventive concept as defined by the appended claims and their equivalents.

WHAT IS CLAIMED IS:

1. An apparatus for coating a solid particle with a coating material, comprising:
  - (a) a first chamber;
  - 5 (b) a flow restrictor disposed at one end of the chamber;
  - (c) a liquid inlet line disposed in fluid communication with the chamber for metering a liquid composition comprising a coating material into the
  - 10 first chamber and through the flow restrictor;
  - (d) a gas inlet line disposed in fluid communication with the first chamber for injecting a first gas stream through the flow restrictor to create a zone of turbulence; and
  - 15 (e) means disposed in the gas inlet line and upstream of the flow restrictor for heating the first gas stream prior to injection through the flow restrictor.
2. The apparatus of claim 1, wherein the liquid
- 20 inlet line is positioned in the flow restrictor.
3. The apparatus of claim 1, wherein the flow restrictor has an outlet end, and the outlet end is positioned in the first chamber.
4. The apparatus of claim 1, further including a
- 25 hopper for introducing a solid particle to the zone of turbulence.
5. The apparatus of claim 4, wherein the flow restrictor has an outlet end, and the outlet end is positioned in the first chamber beneath the hopper.
- 30 6. The apparatus of claim 4, wherein the flow restrictor has an outlet end, and the outlet end is positioned in the first chamber beneath the center line of the hopper.
7. The apparatus of claim 1, further comprising
- 35 a second chamber surrounding the first chamber.
8. The apparatus of claim 7, wherein the axial length of the zone of turbulence is about ten times the diameter of the second chamber.

9. The apparatus of claim 7, wherein the second chamber has an inlet for introducing a second gas stream having a pressure lower than the pressure of the first gas stream into the second chamber.

5 10. A process for coating a solid particle with a coating material, comprising the steps of:

(a) metering a liquid composition comprising the coating material into a flow restrictor;

10 (b) injecting a gas stream through the flow restrictor concurrently with step (a) to create a zone of turbulence at the outlet of the flow restrictor, thereby atomizing the liquid composition;

(c) heating the gas stream prior to injecting the gas stream through the flow restrictor;  
15 and

(d) adding a solid particle to the zone of turbulence concurrently with steps (a) and (b) to mix the solid particle with the atomized liquid composition, wherein the mixing at the zone turbulence coats the solid particle the coating material.  
20

11. The process of claim 10, wherein the liquid composition comprises a liquid and a solid.

12. The process of claim 11, wherein the solid is dissolved in the liquid.

25 13. The process of claim 11, wherein the solid is undissolved in the liquid.

14. The process of any of claims 11-13, wherein the gas stream is heated to a temperature sufficient to vaporize the liquid and to leave the solid remaining and the mixing at the zone of turbulence coats the solid particle with the remaining solid.  
30

15. The process of claim 16, wherein the liquid composition comprises a melt.

16. The process of claim 15, wherein the mixing at the zone of turbulence coats the solid particle with the melt.  
35

17. The process of claim 10, further comprising the step of adding another gas stream upstream of the

zone of turbulence for cooling and conveying the coated solid particles.

18. The process of claim 10, wherein the step of injecting the heated gas stream further comprises  
5 accelerating the gas stream through the flow restrictor to at least one half the velocity of sound prior to entering the flow restrictor.

19. The process of claim 10, wherein the average residence time of the solid particle within the zone of  
10 turbulence is less than 250 milli-seconds.

20. The process of claim 19, wherein the average residence time is in the range of 25 to 250 milli-seconds.

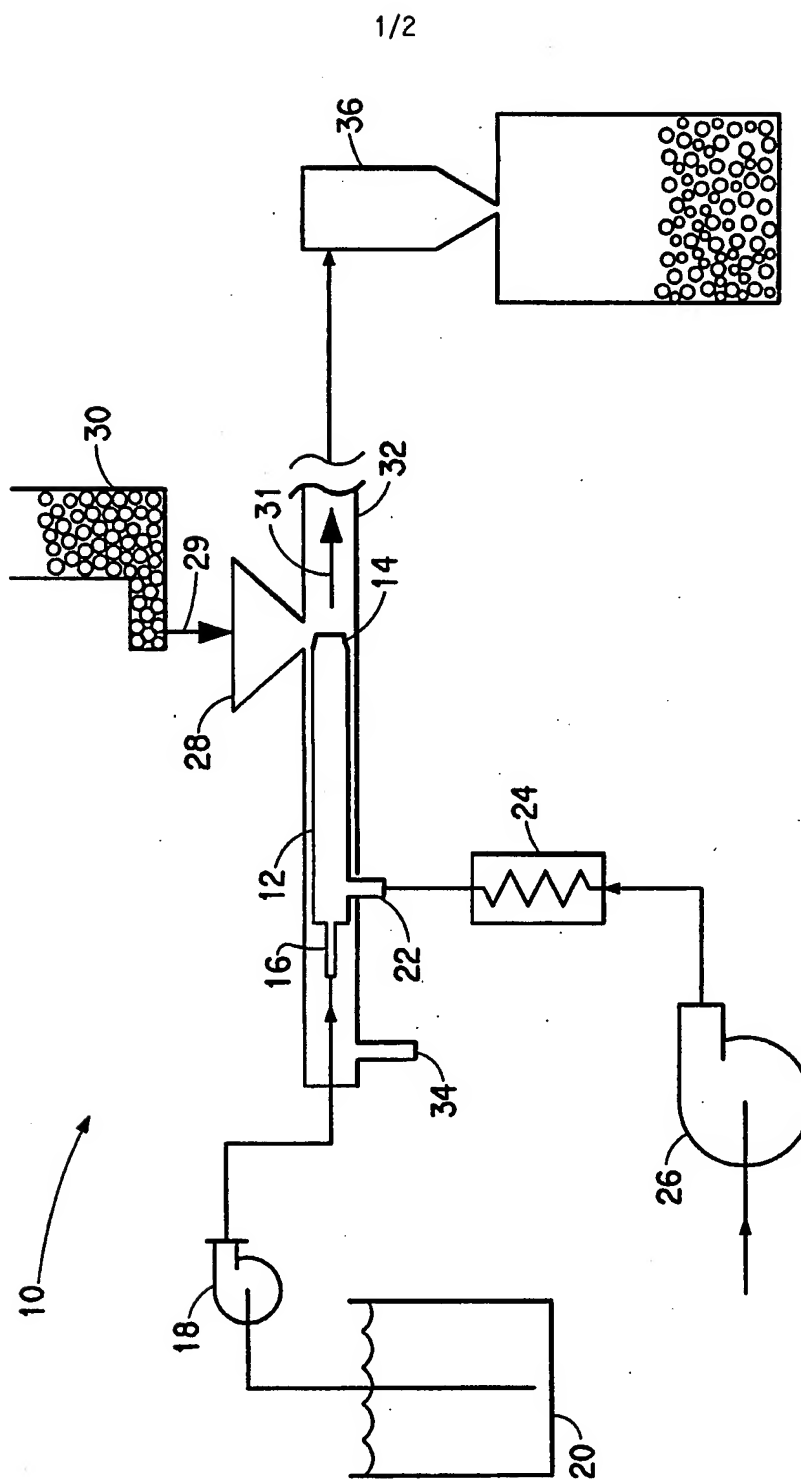
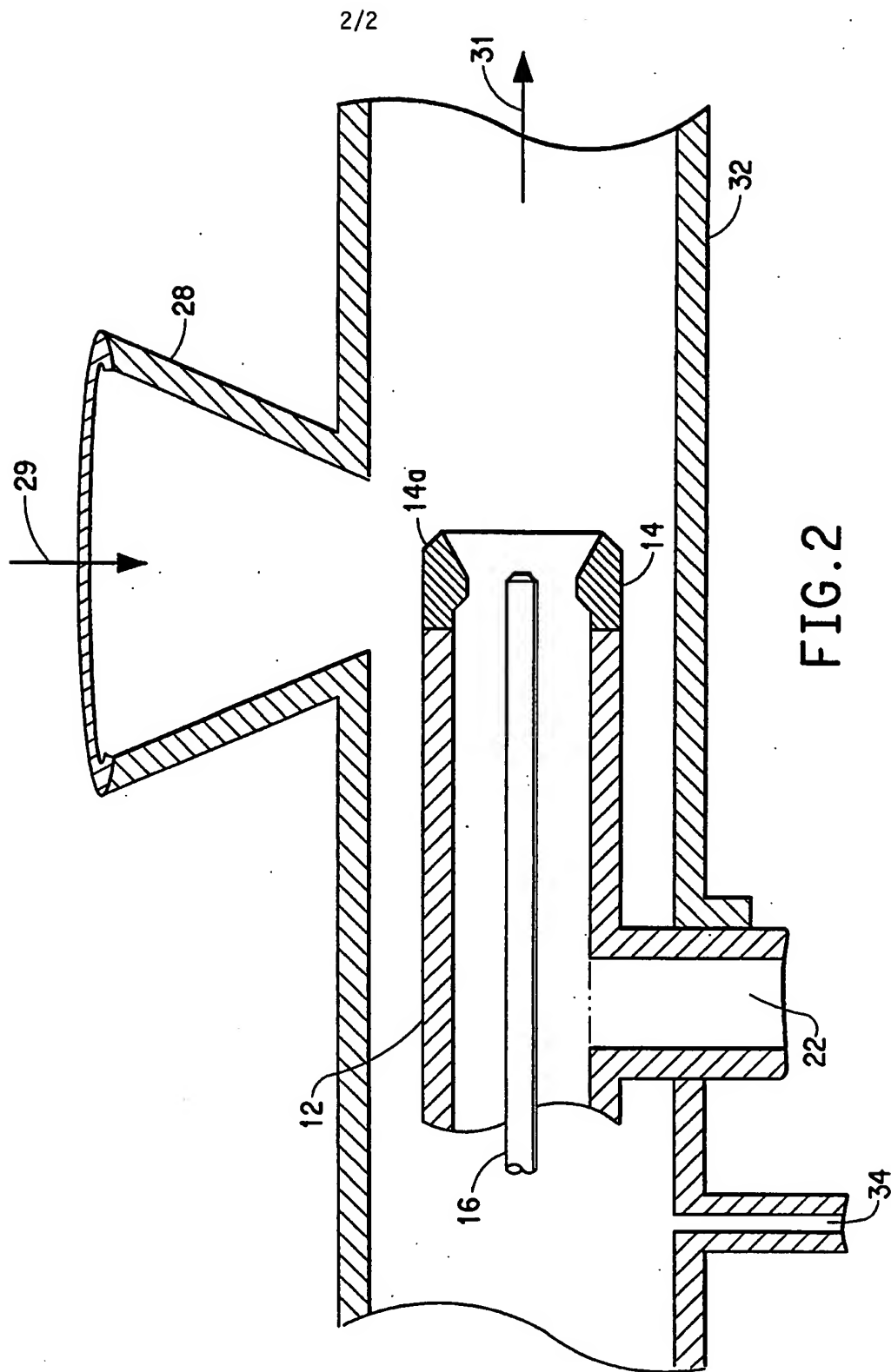


FIG. 1





# INTERNATIONAL SEARCH REPORT

Int. Application No  
PCT/US 96/13582

**A. CLASSIFICATION OF SUBJECT MATTER**  
IPC 6 B01J2/00

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)  
IPC 6 B01J A23G C04B C01B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US,A,3 704 786 (MARC LERNER) 5 December 1972  see column 6, line 25 - line 67; figure 3 ---	1,3,4,7, 9,10, 15-17
A	US,A,4 592 302 (SHIMESU MOTOYAMA) 3 June 1986  see column 3, line 4 - line 18; figures ---	1,4-6, 10,18
A	DE,A,24 29 630 (LIEDL, G) 16 January 1975  see figure ---	1,10
A	EP,A,0 435 426 (FUKUJI CHEMICAL INDUSTRY CO. LTD.) 3 July 1991  see figures 1,3 -----	1,10

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

\* Special categories of cited documents :

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
- \*E\* earlier document but published on or after the international filing date
- \*L\* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- \*O\* document referring to an oral disclosure, use, exhibition or other means
- \*P\* document published prior to the international filing date but later than the priority date claimed

- \*T\* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- \*X\* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- \*Y\* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
- \*&\* document member of the same patent family

Date of the actual completion of the international search

3 December 1996

Date of mailing of the international search report

18.12.96

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax (+31-70) 340-3016

Authorized officer

Van Belleghem, W

# INTERNATIONAL SEARCH REPORT

Information on patent family members

Inte:     nal Application No

PCT/US 96/13582

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US-A-3704786	05-12-72	NONE	
US-A-4592302	03-06-86	NONE	
DE-A-2429630	16-01-75	JP-C- 946690	30-03-79
		JP-A- 50018325	26-02-75
		JP-B- 53028030	11-08-78
		GB-A- 1470227	14-04-77
EP-A-435426	03-07-91	JP-A- 3202137	03-09-91
		DE-T- 69002098	07-10-93
		US-A- 5188868	23-02-93